

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

COMPLETION OF FILING NATIONAL PHASE OF PCT APPLICATION
UNDER RULE 35 USC 371 AND 37 CFR 1.494(C) OR 1.495(C)**BOX PCT****COMPLETION**
For PCT Cases Only

Attn: Application Division

In re PATENT APPLICATION ofInventor(s): BERRY ET ALAppln. No.: 09 600,690
Series Code ☐ Serial No. ☐Atty. Dkt. PM 271641 C382.04/U
M# Client Ref

National Phase Field

Based on PCT GB99 00145
☐ Country Code & YearTitle: POLYSACCHARIDE CONJUGATES CAPABLE OF BINDING
TO CELLULOSE

Date: September 11, 2000

FILING OF ITEM(S) LATE IN PCT/USA NATIONAL CASEAsst. Commissioner of Patents
Washington, DC 20231

Sir:

The following completes the filing of the subject application under Rule 494(c)/495(c). Please accept the following attached items:

1. Missing Requirements Notice (PCT/DO/EO/905) ☒ copy attached ☐ not yet received
2. ☒ **Signed Declaration** ☐ Original ☒ Facsimile/Copy ☐ with spec/claims attached
3. ☐ **Translation** of the International Application into English including:
 - a. ☐ Request;
 - b. ☐ Abstract
 - c. ☐ pgs. Spec. and Claims;
 - d. ☐ Translation verification
 - e. ☐ sheets Drawing which are: ☐ informal ☐ formal of size ☐ A4 ☐ 11"
4. ☐ Copy of **International Search Report (ISR)** attached (☐ page(s))
 - a. ☐ plus Annex of family members (☐ page(s))
5. **Information Disclosure Statement** including
 - a. ☐ From PTO-1449 listing documents
 - b. ☐ Copies of document(s) listed on Form PTO-1449
 - c. ☐ A concise explanation of ISR references is given in the ISR
6. ☒ **Assignment** and cover sheet. Please return the recorded assignment to the undersigned.
7. ☐ Copy of Power to international application agent.
8. ☐ (No.) Small Entity Statement(s) establishing "small entity" status under Rules 9 & 27.
9. ☐ Formal Drawings: ☐ sheet(s) ☐ informal; ☐ formal of size: ☐ A4 ☐ 11"
10. ☒ Please immediately start national examination procedures (35 USC 371(f))

09/13/2000 ERINAND0 0000067 09600690

01 FC:968 260.00 DP
02 FC:154 130.00 DP
Documents

11. ☐ Attached:
12. ☐ Preliminary Amendment:
13. ☒ Basic U.S. National fee per Rule 492(a)(1)-(4) was previously timely filed.:
14. **Calculation of remaining fees due (if any):** based on amended claim(s) per above item
☐ 12 (above) or item(s) in PAT-112 (filed previously) ☐ 12 ☐ 14 ☐ 17 ☐ 25
15. **CLAIMS FEES** ☐ previously paid ☒ paid herewith as follows:
- 15A. Small Entity Statement ☐ Herewith ☐ Previously Filed

				Large/Small Entity		Fee Code
16. Total Effective Claims	14	minus 20 =	0	x \$18/\$9	+0	966/967
17. Independent Claims	1	minus 3 =	0	x \$78/\$39	+0	964/965
18. If any proper multiple dependent claim (ignore improper) is present,				\$260/\$130	+260	968/969
19. Filing Declaration late, fee paid <input type="checkbox"/> previously <input checked="" type="checkbox"/> now				\$130/\$65	+130	154/254
20. SUBTOTAL					\$390	
21. Original due date: September 9, 2000 (Saturday)						
22. Petition is hereby made to extend the original due date to				(1 mo)	\$110/\$55 =	+
cover the date this response is filed for which the requisite fee				(2mos)	\$380/\$190 =	
is attached				(3mos)	\$870/\$435 =	
				(4mos)	\$1360/\$680 =	
23. If "non-English" box 3 is X'd, add Rule 17(k) processing fee				\$130	+0	139
24. If "assignment" box 6 is X'd, add recording fee				\$40	+40	581
25. TOTAL FEE ENCLOSED =					\$430	

(Our Deposit Account No. 03-3975)

(Our Order No. 41301 271641

C#

M#

CHARGE STATEMENT: The Commissioner is hereby authorized to charge any fee specifically authorized hereafter, or any missing or insufficient fee(s) filed, or asserted to be filed, or which should have been filed herewith or concerning any paper filed hereafter, and which may be required under Rules 16-18 (missing or insufficient fee only) now or hereafter relative to this application and the resulting Official document under Rule 20, or credit any overpayment, to our Account/Order Nos. shown above for which purpose a duplicate copy of this sheet is attached.

This CHARGE STATEMENT does not authorize charge of the issue fee until/unless an issue fee transmittal form is filed.

Pillsbury Madison & Sutro LLP
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NOTE: File in duplicate with PTO receipt (PAT-103A) and attachments

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
REQUEST FOR FILING NATIONAL PHASE OF
PCT APPLICATION UNDER 35 U.S.C. 371 AND 37 CFR 1.494 OR 1.495

Commissioner of Patents
and Trademarks
Washington, D.C. 20231

(Our Deposit Account No. 03-3975)

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)

Atty Dkt: PM 271641 /HCM/UK/C382.04/
M# /Client Ref.

From: Pillsbury Madison & Sutro LLP, IP Group:

Date: July 14, 2000

This is a **REQUEST** for **FILING** a PCT/USA National Phase Application based on:

- | | | |
|---|---|---|
| 1. International Application
<u>PCT/GB99/00145</u>
↑ country code | 2. International Filing Date
15 January 1999
Day MONTH Year | 3. Earliest Priority Date Claimed
16 January 1998
Day MONTH Year
(use item 2 if no earlier priority) |
|---|---|---|

4. Measured from the earliest priority date in item 3, this PCT/USA National Phase Application Request is being filed within:

(a) ☐ 20 months from above item 3 date (b) ☒ 30 months from above item 3 date,

(c) Therefore, the due date (unextendable) is July 16, 2000

5. Title of Invention POLYSACCHARIDE CONJUGATES CAPABLE OF BINDING TO CELLULOSE

6. Inventor(s) BERRY, Mark John et al

Applicant herewith submits the following under 35 U.S.C. 371 to effect filing:

7. ☒ Please immediately start national examination procedures (35 U.S.C. 371 (f)).
8. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)) is transmitted herewith (file if in English but, if in foreign language, file only if not transmitted to PTO by the International Bureau) including:
- a. ☒ Request;
 - b. ☒ Abstract;
 - c. 11 pgs. Spec. and Claims;
 - d. _____ sheet(s) Drawing which are ☐ informal ☐ formal of size ☐ A4 ☐ 11"
9. ☒ A copy of the International Application has been transmitted by the International Bureau.
10. A translation of the International Application into English (35 U.S.C. 371(c)(2))
- a. ☐ is transmitted herewith including: (1) ☐ Request; (2) ☐ Abstract;
 - (3) _____ pgs. Spec. and Claims;
 - (4) _____ sheet(s) Drawing which are: ☐ informal ☐ formal of size ☐ A4 ☐ 11"
 - b. ☐ is not required, as the application was filed in English.
 - c. ☐ is not herewith, but will be filed when required by the forthcoming PTO Missing Requirements Notice per Rule 494(c) if box 4(a) is X'd or Rule 495(c) if box 4(b) is X'd.
 - d. ☐ Translation verification attached (not required now).

RE: USA National Filing of PCT/GB99/00145

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11. ☒ **PLEASE AMEND** the specification before its first line by inserting as a separate paragraph:
 a. ☒ -This application is the national phase of international application PCT/GB99/00145
 filed January 15, 1999 which designated the U.S.--
 b. ☐ -This application also claims the benefit of U.S. Provisional Application No. 60/____, filed ____.
12. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)), i.e., **before 18th month from first priority date above in item 3, are transmitted herewith (file only if in English) including:**
13. ☒ PCT Article 19 claim amendments (if any) have been transmitted by the International Bureau
14. ☐ Translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)), i.e., of **claim amendments made before 18th month, is attached (required by 20th month from the date in item 3 if box 4(a) above is X'd, or 30th month if box 4(b) is X'd, or else amendments will be considered canceled).**
15. **A declaration of the inventor (35 U.S.C. 371(c)(4))**
 a. ☐ is submitted herewith ☐ Original ☐ Facsimile/Copy
 b. ☒ is not herewith, but will be filed when required by the forthcoming PTO Missing Requirements Notice per Rule 494(c) if box 4(a) is X'd or Rule 495(c) if box 4(b) is X'd.
16. **An International Search Report (ISR):**
 a. Was prepared by ☒ European Patent Office ☐ Japanese Patent Office ☐ Other
 b. ☒ has been transmitted by the international Bureau to PTO.
 c. ☒ copy herewith (2 pg(s).) ☒ plus Annex of family members (1 pg(s)).
17. **International Preliminary Examination Report (IPER):**
 a. ☒ has been transmitted (if this letter is filed after 28 months from date in item 3) in English by the International Bureau with Annexes (if any) in original language.
 b. ☒ copy herewith in English.
 c.1 ☐ IPER Annex(es) in original language ("Annexes" are amendments made to claims/spec/drawings during Examination) including attached amended:
 c.2 ☐ Specification/claim pages #____ claims #____
 Dwg Sheets #____
 d. ☐ Translation of Annex(es) to IPER **(required by 30th month due date, or else annexed amendments will be considered canceled).**
18. **Information Disclosure Statement** including:
 a. ☒ Attached Form PTO-1449 listing documents
 b. ☒ Attached copies of documents listed on Form PTO-1449
 c. ☐ A concise explanation of relevance of ISR references is given in the ISR.
19. ☐ **Assignment** document and Cover Sheet for recording are attached. Please mail the recorded assignment document back to the person whose signature, name and address appear at the end of this letter.
20. ☐ Copy of Power to IA agent.
21. ☐ **Drawings** (complete only if 8d or 10a(4) not completed): ____ sheet(s) per set: ☐ 1 set informal;
☐ Formal of size ☐ A4 ☐ 11"
22. ☐ ____ (No.) **Verified Statement(s)** establishing "small entity" status under Rules 9 & 27
23. **Priority** is hereby claimed under 35 U.S.C. 119/365 based on the priority claim and the certified copy, both filed in the International Application during the international stage based on the filing in (country) EUROPE of:
Application No. Filing Date Application No. Filing Date
 (1) 98300291.6 January 16, 1998 (2) _____
 (3) _____ (4) _____
 (5) _____ (6) _____
- a. ☒ See Form PCT/IB/304 sent to US/DO with copy of priority documents. If copy has not been received, **please proceed promptly to obtain same from the IB.**
 b. ☒ Copy of Form PCT/IB/304 attached.

534 Rec'd PCT/PT 14 JUL 2000

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POLYSACCHARIDE CONJUGATES CAPABLE OF BINDING TO CELLULOSE

Field of Invention

- This invention relates to binding of polysaccharides and concerns a cellulose-binding polysaccharide conjugate, products including the polysaccharide conjugate, and targeting methods using the polysaccharide conjugate. In the context of the invention the term "polysaccharide" is intended to cover polysaccharides and oligosaccharides, and references to "polysaccharide" and "polysaccharide conjugate" should be construed accordingly.

Background to the Invention

- 10 It is known that various naturally occurring polysaccharides such as pea xyloglucan, tamarind seed xyloglucan, etc bind to cellulose by a polysaccharide:polysaccharide interaction; indeed this binding ability is important in the functioning of plant cell walls.
- The paper by Hayashi et al entitled "Pea Xyloglucan and Cellulose" in Plant Physiol. (1987) 83, 384-389 describes investigations of binding of pea xyloglucan to cellulose, using fluorescein-labelled xyloglucan prepared by treating xyloglucan with cyanogen bromide (CNBr) and incubating with fluoresceinamine, and also using radiiodinated xyloglucan prepared by reaction of ^{125}I with the fluorescein moiety on xyloglucan. These labels were used to trace the binding of the polysaccharide and are among the smallest molecular label entities known.
- 15 The present invention is based on the surprising discovery that polysaccharides with much larger attached entities than those used by Hayashi et al can still bind rapidly with high efficiency to cellulose by polysaccharide:polysaccharide interaction. This is surprising because binding occurs at multiple sites along the backbones of the polysaccharides, rather than at a single binding site as with antibody-antigen interactions, and it would have been predicted that binding would have been disrupted by the attachment of large entities to cellulose-binding polysaccharides. The invention thus opens up the possibility of using polysaccharides to target attached particles to cellulose, eg in fabric, paper, etc.
- 20
- 25

Summary of the invention

In one aspect the present invention provides a polysaccharide conjugate comprising a polysaccharide attached to a particle carrying perfume, the polysaccharide conjugate being capable of binding to cellulose.

- 5 The polysaccharide may be one that binds naturally to cellulose or has been derivatised or otherwise modified to bind to cellulose. The polysaccharide may be naturally occurring or synthetic.

- The polysaccharide desirably has a 1-4 linked β glycan (generalised sugar) backbone structure, which is stereochemically compatible with cellulose, such as a glucan backbone (consisting of β 1-4 linked glucose residues), a mannan backbone (consisting of β 1-4 linked mannose residues) or a xylan backbone (consisting of β 1-4 linked xylose residues). Suitable polysaccharides include xyloglucans, glucomannans, mannans, galactomannans, β (1-3), (1-4) glucan and the xylan family incorporating glucurono-, arabino- and glucuronoarabinoxylan. See "Physiology and Biochemistry of Plant Cell Walls" (1990) by C Brett and K Waldron for a discussion of these materials.

- The minimum chain length requirement for cellulose oligomers to bind to cellulose is 4 glucose units. For xyloglucans, the side chains make the binding less efficient and 12 backbone glucose units (ie. about 25 total sugar units) are required for binding to cellulose. Structural considerations suggest galactomannans are intermediate in binding efficiency, and about 6 to 8 backbone residues are expected to be required for binding to cellulose. The polysaccharide should thus have at least 4, and preferably at least 10, backbone residues, which are preferably β 1-4 linked.

- Naturally occurring polysaccharides that bind rapidly and strongly to cellulose by polysaccharide:polysaccharide interaction include xyloglucans such as pea xyloglucan and tamarind seed xyloglucan (TXG) (which has a β 1-4 linked glucan backbone with side chains of α -D xylopyranose and β -D-galactopyranosyl-(1-2)- α -D-xylo-pyranose, both 1-6 linked to the backbone; see Gidley et al Carbohydrate Research, 214 (1991) 200-314 for a discussion of the structure of tamarind seed polysaccharide); and galactomannans, particularly low galactose galactomannans, such as locust bean gum (LBG) (which has a mannan backbone of β 1-4 linked mannose residues, with single unit galactose side chains linked α 1-6 to the backbone), enzyme modified guar (EMG) (guar gum has the same structural units as LBG but has a much higher level of galactose substitution, to the extent that there is not enough accessible mannan backbone through which to bind cellulose. EMG is produced by enzymic removal from guar gum of a controllable

- percentage of the galactose residues to produce a range of materials that are capable of binding to cellulose, but are cheaper and more consistently available than LBG. See Bulpin et al Carbohydrate Polymers 12 (1990) 155-168 for a discussion of EMG), tara galactomannan and cassia galactomannan. These materials are commercially available and thus provide potentially useful sources of suitable polysaccharides. These materials have the advantages of being relatively cheap, and already being accepted for food use.

The polysaccharide desirably has side chain galactose residues susceptible to oxidation by galactose oxidase, for production of an aldehyde group for coupling purposes, as will be described below. TXG, LBG and EMG have such galactose residues.

- 10 The particle may be of a range of materials including silica, particular porous silica, organic polymer etc.

The perfume may be carried by the particle in a variety of ways, including absorption, adsorption, impregnation and encapsulation.

- 15 Absorption of a perfume by particles can be brought about simply by bringing the perfume and the particles into contact, and allowing them to stand. The perfume molecules can enter the particles by diffusion.

An alternative to the use of solid particles is to form hollow capsules in which a shell encapsulates the perfume.

- 20 One approach to the preparation of microcapsules of perfume is to disperse droplets of the perfume in an aqueous phase which contains water soluble polymer, and then form a polymer shell around these perfume droplets by coacervation of the polymer at the interface between the perfume and the aqueous phase. Once formed, the capsule wall usually requires further treatment to strengthen it. The encapsulation of perfume by coacervation has been described by Meyer, A in Chimica, 46, 101 (1992) and in US-A-5051305.

A second approach to the formation of microcapsules of perfume is to disperse perfume droplets in an aqueous phase, and then bring about a polymerisation reaction at the interface between the droplets and the aqueous phase. The polymerisation reaction which has mostly been employed is the formation of an aminoplast resin. This has been used for

perfume encapsulation, as disclosed in US-A-4681806. A typical procedure for the production of aminoplast resin capsules enclosing perfume is set out in US-A-4234627, which refers back to US-A-3516941.

- 5 A further possibility is to form solid polymer particles, absorb the perfume into these, and then encapsulate these particles.

- 10 Further information on encapsulation techniques is given in Risch, S.J., Reineccius, G.A. (Ed), "Encapsulation and controlled release of food ingredients", ACS symposium series 590, Washington DC, 1995. It is to be noted that not all of the encapsulation techniques described in this reference are necessarily suitable for the preparation of particles for use in this invention. For instance, spray drying, which is the most widely used encapsulation technique, generally produces water-soluble particles which may not be particularly suitable. However, the person skilled in the art will readily be able to select suitable techniques.

- 15 The perfume typically constitutes between 1 and 90% of the total weight of the particle, and preferably constitutes at least 5% by weight to be commercially attractive. Using encapsulation techniques, perfume loadings of up to about 70% can be achieved, while absorption techniques, eg using highly absorbing silicas, can achieve loadings of up to about 90%.

- 20 The nature of the perfume is not critical to the invention. As is well known, a perfume normally consists of a mixture of a number of perfumery materials, each of which has a fragrance. The number of perfumery materials in a perfume is typically ten or more. The range of fragrant materials used in perfumery is very wide; the materials come from a variety of chemical classes, but in general are water-insoluble oils. In many instances, the molecular weight of a perfumery material is in excess of 150, but does not exceed 300.

- 25 The perfumes used in the present invention can be mixtures of conventional perfumery materials. Perfumery materials which may be used include: acetyl cedrene, 4-acetoxy-3-pentyltetrahydropyran, 4-acetyl-6-t-butyl-1,1-dimethylindane, available under the trademark "CELESTOLIDE", 5-acetyl-1,1,2,3,3,6-hexamethylindane, available under the trademark "PHANTOLIDE", 6-acetyl-1-isopropyl-2,3,3,5-tetramethylindane, available under the trademark "TRASEOLIDE", alpha-n-amylocinnamic aldehyde, amyl salicylate, aubepine, aubepine nitrile, aurantion, 2-t-butylcyclohexyl acetate, 2-t-butylcyclohexanol,
- 30

- 3-(p-t-butylphenyl)propanal, 4-t-butylcyclohexyl acetate, 4-t-butyl-3,5-dinitro-2,6-dimethyl acetophenone, 4-t-butylcyclohexanol, benzoin siam resinoids, benzyl benzoate, benzyl acetate, benzyl propionate, benzyl salicylate, benzyl isoamyl ether, benzyl alcohol, bergamot oil, bornyl acetate, butyl salicylate, carvacrol, cedar atlas oil, cedryl methyl ether, cedryl acetate, cinnamic alcohol, cinnamyl propionate, cis-3-hexenol, cis-3-hexenyl salicylate, citronella oil, citronellol, citronellonitrile, citronellyl acetate,
- 5 citronellyloxyacetaldehyde, cloveleaf oil, coumarin, 9-decen-1-ol, n-decanal, n-dodecanal, decanol, decyl acetate, diethyl phthalate, dihydromyrcenol, dihydromyrcenyl formate, dihydromyrcenyl acetate, dihydroterpinyl acetate, dimethylbenzyl carbonyl acetate,
- 10 dimethylbenzylcarbinol, dimethylheptanol, dimethyloctanol, dimyrcetol, diphenyl oxide, ethyl naphthyl ether, ethyl vanillin, ethylene brassylate, eugenol, florocyclene, geraniol, geranium oil, geranonitrile, geranyl nitrile, geranyl acetate, 1,1,2,4,4,7-hexamethyl-6-acetyl-1,2,3,4-tetrahydronaphthalene, available under the trademark "TONALID", 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta-
- 15 2-benzopyran, available under the trade mark "GALAXOLIDE", 2-n-heptylcyclopentanone, 3a,4,5,6,7,7a-hexahydro-4,7-methano-1(3)H-inden-6-ylpropionate, available under the trademark "FLOROCYCLEN", 3a-4,5,6,7,7a-hexahydro-4,7-methano-1(3)H-inden-6-ylacetate, available under the trademark "JASMACYCLEN", 4-(4'-hydroxy-4'-methylpentyl)-3-cyclohexenecarbaldehyde, alpha-hexylcinammic
- 20 aldehyde, heliotropin, Hercolyn D, hexyl aldol, hexyl cinnamic aldehyde, hexyl salicylate, hydroxycitronellal, i-nonyl formate, 3-isocamphylcyclohexanol, 4-isopropylcyclohexanol, 4-isopropylcyclohexyl methanol, indole, ionones, irones, isoamyl salicylate, isoborneol, isobornyl acetate, isobutyl salicylate, isobutylbenzoate, isobutylphenyl acetate, isoeugenol, isolongifolone, isomethyl ionones, isononanol, isononyl acetate, isopulegol, lavandin oil,
- 25 lemongrass oil, linalool, linalyl acetate, LRG 201, 1-menthol, 2-methyl-3-(p-isopropylphenyl)propanal, 2-methyl-3-(p-t-butylphenyl)propanal, 3-methyl-2-pentyl-cyclopentanone, 3-methyl-5-phenyl-pentanol, alpha and beta methyl naphthyl ketones, methyl ionones, methyl dihydrojasmonate, methyl naphthyl ether, methyl 4-propyl phenyl ether, Mousse de chene Yugo, Musk ambrette, myrtenol, neroli oil,
- 30 nonanediol-1,3-diacetate, nonanol, nonanolide-1,4, nopol acetate, 1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethyl-2-acetyl-naphthalene, available under the trademark "ISO-E-SUPER", octanol, Oppoponax resinoid, orange oil, p-t-amylicyclohexanone, p-t-butylmethylhydrocinnamic aldehyde, 2-phenylethanol, 2-phenylethyl acetate, 2-phenylpropanol, 3-phenylpropanol, para-menthan-7-ol,
- 35 para-t-butylphenyl methyl ether, patchouli oil, pelargone, petitgrain oil, phenoxethyl isobutyrate, phenylacetaldehyde diethyl acetal, phenylacetaldehyde dimethyl acetal,

- phenylethyl n-butyl ether, phenylethyl isoamyl ether, phenylethylphenyl acetate, pimento leaf oil, rose-d-oxide, Sandalone, styrallyl acetate, 1,1,4,4-tetramethyl-6-acetyl-7-ethyl-1,2,3,4-tetrahydronaphthalene, available under the trademark "VERSALIDE", 3,3,5-trimethyl hexyl acetate, 3,5,5-trimethylcyclohexanol, terpineol, terpinyl acetate,
- 5 tetrahydrogeraniol, tetrahydrolinalool, tetrahydromuguol, tetrahydromyrcenol, thyme oil, trichloromethylphenylcarbiny acetate, tricyclodeceny acetate, tricyclodeceny propionate, 10-undecen-1-al, gamma undecalactone, 10-undecen-1-ol, undecanol, vanillin, vetiverol, vetiveryl acetate, vetyvert oil, acetate and propionate esters of alcohol in the list above, aromatic nitromusk fragrances, indane musk fragrances, isochroman musk fragrances,
- 10 macrocyclic ketones, macrolactone musk fragrances, and tetralin musk fragrances.

The perfume may be present in known manner in solvents or diluents, such as ethanol, isopropanol, diethylene glycol monoethyl ether, dipropylene glycol, diethyl phthalate and triethyl citrate.

- Perfumes which are used in this invention may, if desired, have deodorant properties, for example, as disclosed in US-A- 4303679, US-A-4663068 and EP-A-545556.
- 15

If the particles are solid particles which are impregnated with perfume after manufacture, the absorption of perfume can be enhanced by choosing perfumery materials with a hydrophobic character or mixing a hydrophobic oil into the perfume. Examples of hydrophobic oils which can enhance perfume uptake are dibutylphthalate, alkane mixtures such as isoparaffin and di (C₈-C₁₀ alkyl) propylene glycol diester.

20

The particles suitably have a diameter in the range 0.5 to 100 microns. The lower end of this range (0.5 to 5 microns) covers small colloidal particles and molecular complexes.

The particle may be attached to the polysaccharide by a range of physical or chemical means.

- 25 Polysaccharide is conveniently physically attached to particles, eg by adsorption. For example, porous silica particles have surface properties that enable firm adsorption of polysaccharide.

Chemical attachment techniques may also be used. For particles carrying surface amino groups, attachment can be by a number of techniques. For example, amino groups are

- conveniently chemically linked to polysaccharides having galactose side chains by enzymically oxidising the galactose, eg using galactose oxidase, to produce an aldehyde group to which an amino group of a particle can be chemically linked. As noted above, TXG, LBG and EMG have suitable galactose side chains. For polysaccharides not having
- 5 suitable galactose side chains, different methods of chemical linking of amino groups can be used. Alternative techniques include limited periodate oxidation, which requires the polysaccharide to have two adjacent hydroxyl groups in cis orientation, and results in the production of aldehyde groups which can be reductively aminated. A further possibility is
- 10 reaction with cyanogen bromide (CNBr) which inserts into sugar rings at vicinal diols, both in the backbone and side chains, to provide an isourea linkage to amino groups. It is preferred to use chemical techniques that do not affect the polysaccharide backbone length, which would reduce the cellulose-binding capability of the polysaccharide. For particles with surface carboxyl or hydroxyl groups, other known forms of chemical linkage may be used. As a further possibility, where the particle is a liposome or micelle,
- 15 hydrophobic tails fixed to the polysaccharide can be inserted therein.

Because the polysaccharide conjugate binds to cellulose, which is present in cotton and other fabrics, paper, etc. binding of the conjugate to cellulose brings the attached particle carrying perfume into close proximity to a surface of or containing cellulose. The invention thus enables targeting of perfume-bearing particles to such surfaces.

- 20 This targeting function finds particular practical applications, in targeting of particles containing perfume to bind to fabric. The perfume may be adsorbed, impregnated or encapsulated in the particle. Polysaccharide-particle conjugates of this sort thus find use as ingredients in laundry products, such as fabric washing products and fabric conditioning products, and also personal products, eg. for targeting perfume to bind to clothes. Another
- 25 potential use is in paper products, such as disposable paper wipes.

In a preferred embodiment, the particles are porous and contain perfume in the pores. This embodiment involves filling the pores of the particles with the perfume and then blocking the pores with a coating of the polysaccharide so that the perfume does not come out of the particle again easily. It may, however, be possible to effect perfume release if

30 desirable, eg by ironing. Moreover, the coating has the combined effect of sealing the perfume in the pores and giving the particles a specific affinity for cellulose or cellulose-containing surfaces.

The invention may also be used for targeting perfume-bearing particles to bind paper to produce perfumed paper.

An additional benefit of the invention arises from the fact that, unlike most other targeting molecules, cellulose-binding polysaccharides are especially robust. Proteins such as cellulose binding domain can be inactivated (denatured) by heat or aggressive surfactants, while polysaccharides such as LBG, TXG, etc are completely unaffected by such treatments. The polysaccharide conjugates of the invention thus offer the considerable advantage of extra stability and product compatibility compared with other targeting molecules.

- 10 In a further aspect, the present invention provides a product incorporating a polysaccharide conjugate in accordance with the invention.

The product is conveniently a laundry product such as a fabric washing product, eg a detergent product, or a fabric conditioning product.

- 15 The invention also finds application in personal products, eg for targeting perfume to bind to clothes.

The product may otherwise be of generally conventional formulation, as is well known to those skilled in the art. For a discussion of known detergent compositions see, for example, WO 95/34628, particularly pages 11 to 15.

- 20 The present invention also provides a method of targeting binding of a particle carrying perfume to cellulose by use of a polysaccharide conjugate in accordance with the invention.

The invention will be further described by way of illustration, in the following Example.

EXAMPLE 1

Binding of Locust Bean gum (LBG) to fragrance-enriched particles for targeting to fabric.

- 25 MATERIALS AND METHODS

Preparation of particles

Porous silica particles (mean pore-size 2nm, average particle size 9 μ m) were obtained from Joseph Crosfield & Sons (Warrington U.K.). Approximately 25 mgs of silica was placed in each of two round-bottom plastics tubes. 0.5mls of the fragrance florocyclene (obtained from Quest International, Ashford U.K.) was added to each of the tubes and mixed thoroughly. The lids of the tubes were sealed to prevent evaporation of the fragrance.

The two tubes were rotated overnight with a gentle tumbling motion to allow the fragrance molecules to enter the pores of the particles. This was done at ambient temperature.

The tubes were centrifuged for 5 minutes [13,000 r.p.m. in a microcentrifuge (MSE)] and the supernatants discarded. Each of the two tubes then received a different treatment in which 1mls of one of the following was added.

- a) 1mls of 0.1% LBG (Sigma Product No. G-0753)
- b) 1mls of purified water.

The two tubes were mixed thoroughly and then rotated overnight at ambient temperature. After the overnight treatment, the tubes were centrifuged as before and the supernatants removed. The particles were washed once in water and then twice in saline (pH 7). The particles were then resuspended in 1mls saline (pH 7) and left at ambient temperature until required. There were therefore two different slurries, each containing approximately 25 mgs of silica in 1mls of saline. The slurries were designated a) or b) depending on the treatment received as set out above.

Targeting the particles to fabric

Two squares (approximately 1cm x 1cm) were cut from white cotton cloth.

2mls of saline was added to each of two plastics tubes. 100 μ l of slurry a) was added to one; 100 μ l of slurry b) was added to the other. One cotton square was added to each of the two tubes a) and b).

The two tubes were rotated slowly at room temperature for two hours; after which the cotton squares were removed with tweezers. Each square was placed in a separate Petri dish. The lids of the Petri dishes were used to prevent evaporation of the fragrance.

RESULTS

Eight people were asked to compare cotton squares treated with slurry a) and slurry b) and decide which of the cotton squares smells the strongest.

6 out of 8 said that a) was the strongest

5 1 out of 8 said that b) was the strongest

1 out of 8 could not tell the difference.

It was concluded that the cotton square treated with slurry a) had more fragrance on it due to the targeting effect of the LBG.

Claims

1. A polysaccharide conjugate comprising a polysaccharide attached to a particle carrying perfume, the polysaccharide conjugate being capable of binding to cellulose.
2. A conjugate according to claim 1, wherein the polysaccharide has a 1-4 linked β glycan backbone structure.
3. A conjugate according to claim 2, wherein the polysaccharide has a glucan backbone, a mannan backbone or a xylan backbone.
4. A conjugate according to claim 3, wherein the polysaccharide is selected from xyloglucans, glucomannans, mannans, galactomannans, β (1-3), (1-4) glucan and the xylan family incorporating glucurono-, arabino- and glucuronoarabinoxylan.
5. A conjugate according to claim 4, wherein the polysaccharide is selected from xyloglucans such as tamarind seed xyloglucan (TXG) and pea xyloglucan; and galactomannans, particularly low galactose galactomannans, such as locust bean gum (LBG), enzyme modified guar (EMG), tara galactomannan and cassia galactomannan.
6. A conjugate according to any one of the preceding claims, wherein the polysaccharide has side chain galactose residues susceptible to oxidation by galactose oxidase.
7. A conjugate according to any one of the preceding claims, wherein the particle is physically linked to the polysaccharide.
8. A product incorporating a polysaccharide conjugate in accordance with any one of the preceding claims.
9. A product according to claim 8, comprising a laundry product such as a fabric washing product, eg a detergent product, or a fabric conditioning product.
10. A method of targeting binding of a particle carrying perfume to cellulose by use of a polysaccharide conjugate in accordance with any one of claims 1 to 7.

FOR UTILITY/DESIGN
CIP/PCT NATIONAL/PLANT
ORIGINAL/SUBSTITUTE/SUPPLEMENTAL
DECLARATIONS

RULE 63 (37 C.F.R. 1.63)
DECLARATION AND POWER OF ATTORNEY
FOR PATENT APPLICATION

PM & S
FORM

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

C382.04/U

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name, and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the **INVENTION ENTITLED POLYSACCHARIDE CONJUGATES CAPABLE OF BINDING TO CELLULOSE**

the specification of which (CHECK applicable BOX(ES))
X ☐ A ☐ is attached hereto.
BOX(ES) → ☒ B was filed on July 14, 2000 as U.S. Application No. 09/1600,690
→ ☒ C was filed as PCT International Application No. PCT/GB99/00145 on January 15, 1999
and (if applicable to U.S. or PCT application) was amended on _____
I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56. Except as noted below, I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(a) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International Application which designated at least one other country than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate, or PCT International Application, filed by me or my assignee disclosing the subject matter claimed in this application and having a filing date (1) before that of the application on which priority is claimed, or (2) if no priority claimed, before the filing date of this application.

PRIOR FOREIGN APPLICATION(S)

Number	Country	Day/MONTH/Year Filed	Date first Laid-open or Published	Date Patented or Granted	Priority NOT Claimed
98300291.6	EUROPE	16 January 1996			

If more prior foreign applications, X box at bottom and continue on attached page.

Except as noted below, I hereby claim domestic priority benefit under 35 U.S.C. 119(e) or 120 and/or 365(c) of the indicated United States applications listed below and PCT International applications listed above or below and, if this is a continuation-in-part (CIP) application, insofar as the subject matter disclosed and claimed in this application is in addition to that disclosed in such prior applications. I acknowledge the duty to disclose all information known to me to be material to patentability as defined in 37 C.F.R. 1.56 which became available between the filing date of each such prior application and the national or PCT international filing date of this application.

PRIOR U.S. PROVISIONAL, NONPROVISIONAL AND/OR PCT APPLICATION(S)

Application No. (series code/serial no.)	Day/MONTH/Year Filed	Status	Priority NOT Claimed
		pending, abandoned, patented	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

And I hereby appoint Pillsbury Madison & Suto LLP, Intellectual Property Group, 1100 New York Avenue, N.W., Ninth Floor, East Tower, Washington, D.C. 20005-3618, telephone number (202) 861-3000 (to whom all communications are to be directed) and the below-named persons (of the same address) individually and collectively my attorneys to prosecute this application and to transact all business in the Patent and Trademark Office concerning the same and with the resulting patent, and I hereby authorize them to delete names/numbers below of persons no longer with their firm and to act and rely on instructions from and communicate directly with the person(s)/attorney/firm(s) in connection with which first executed this case to them and by whom which I hereby declare that I have consented after full disclosure to be represented unless/until I instruct the above firm and/or a below attorney in writing to the contrary.

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FOR ADDITIONAL INVENTORS, "X" box ☒ and proceed on the attached page to list each additional inventor.

☐ See additional foreign priorities on attached page (incorporated herein by reference).

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(M#)

DECLARATION AND POWER OF ATTORNEY

(continued)

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City		State/Foreign Country		Country of Citizenship	
Post Office Address					
(include Zip Code)					

Rule 56(a) & (b) = 37 C.F.R. 1.56(a) & (b)
PATENT AND TRADEMARK CASES - RULES OF PRACTICE
DUTY OF DISCLOSURE

- (a) ... Each individual associated with the filing and prosecution of a patent application has a duty of candor and good faith in dealing with the [Patent and Trademark] Office, which includes a duty to disclose to the Office all information known to that individual to be material to patentability... (b) information is material to patentability when it is not cumulative and (1) It also establishes by itself, or in combination with other information, a prima facie case of unpatentability of a claim or (2) refutes, or is inconsistent with, a position the applicant takes in: (i) Opposing an argument of unpatentability relied on by the Office, or (ii) Asserting an argument of patentability

PATENT LAWS 35 U.S.C.

§102. Conditions for patentability; novelty and loss of right to patent

A person shall be entitled to a patent unless--

- (a) the invention was known or used by others in this country, or patented or described in a printed publication in this or a foreign country, before the invention thereof by the applicant for patent or
- (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of the application for patent in the United States, or
- (c) he has abandoned the invention, or
- (d) the invention was first patented or caused to be patented, or was the subject of an inventor's certificate, by the applicant or his legal representatives or assigns in a foreign country prior to the date of the application for patent in this country on an application for patent or inventor's certificate filed more than twelve months* before the filing of the application in the United States, or
- (e) the invention was described in a patent granted on an application for patent by another filed in the United States before the invention thereof by the applicant for patent, or on an international application by another who has fulfilled the requirements of paragraphs (1), (2), and (4) of section 371(c) of this title before the invention thereof by the applicant for patent, or
- (f) he did not himself invent the subject matter sought to be patented, or
- (g) before the applicant's invention thereof the invention was made in this country by another who had not abandoned, suppressed, or concealed it. In determining priority of invention there shall be considered not only the respective dates of conception and reduction to practice of the invention, but also the reasonable diligence of one who was first to conceive and last to reduce to practice, from a time prior to conception by the other.

§103. Condition for patentability; non-obvious subject matter

- (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made. . . .
- (c) Subject matter developed by another person, which qualified as prior art only under subsection (f) or (g) of section 102 of this title, shall not preclude patentability under this section where the subject matter and the claimed invention were, at the time the invention was made, owned by the same person or subject to an obligation of assignment to the same person.

* Six months for Design Applications (35 U.S.C. 172).